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Field factor calculation for the study of the relationships between all the 3-wave non-linear optical interactions in uniaxial and biaxial crystals

B Boulanger and G Marnier
Laboratoire de Minéralogie-Cristallographie et Physique Infrarouge, URA CNRS 809, Université de Nancy I, Faculté des Sciences, BP 239, 54506 Vandoeuvre lès Nancy Cédex, France

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Abstract. We propose to establish, by a general treatment, the relationships between all the 3-wave non-linear optical interactions: sum and difference frequency under collinear phase-matching conditions of types I, II and III in negative and positive uniaxial and biaxial crystals. A classification of these interactions is established, based on the field factor: this tensor entity characterizes the polarizations of the different spectral components of the beam interacting with the crystal. The field factor only depends on linear optical properties and is convenient for the evaluation of the 3-wave mixing effective non-linear susceptibility.

1. Introduction

The following criteria are usually considered for the evaluation of the 3-wave non-linear optical properties of the crystals [1, 2]: non-centrosymmetry of the class of orientation symmetry, density, orientation and polarizability of anharmonic chemical bonds and of course transparency at the frequencies concerned and finally phase-matching conditions.

We show in this paper that a crystal which complies with these conditions does not necessarily permit sizeable non-linear mixing. The coupling can also be nil for certain classes of crystalline symmetry when the interacting waves have a particular configuration of polarization.

Then it is essential to calculate the theoretical efficiency of each interaction. It depends on the effective coefficient χ_{eff}, which is defined by the scalar product of the non-linear polarization $P^{\mathrm{NL}}(\omega)$ with the electric field $E(\omega)$ at the circular frequency ω of the considered wave. Thus, the interaction efficiency depends not only on the second-order electric susceptibility $\chi^{(2)}$ but on the polarization of the waves.

The originality of our work lies in the consideration and study of the product $e_i e_j e_k$ of the electric fields components of the three waves relative to χ_{ijk}. We call this product the field factor, written F_{ijk}, which is the element of the tensor, written $\mathbf{F}$, characteristic of the beam interacting with the crystal. The field factor tensor $\mathbf{F}$ depends only on the linear optical properties: dispersions in frequency of the refractive indices and of the birefringences. We establish, by the calculation of the field factor, the relations between the different 3-wave non-linear optical interactions: sum frequency mixing (SFM), difference frequency mixing (DFM), under the different types of phase-matching conditions, in directions of propagation of positive and negative optical sign, in uniaxial and biaxial classes of symmetry. A classification of these interactions is established.

2. Definitions

2.1. Phase-matching relation and configuration of polarization

Our study requires a precise definition of the non-linear optical interactions at three waves [3]: SFM and DFM, collinear phase-matched in uniaxial and biaxial crystals.

2.1.1. The conservation of energy during the interaction imposes the following relation between the circular frequencies of the three waves:

$$\omega_1 + \omega_2 - \omega_3 = 0. \tag{1}$$

By convention, we take:

$$\omega_1 < \omega_2 \ (<\omega_3). \tag{2}$$

In a phase-matching direction $\boldsymbol{u}$, with the spherical coordinates θ and φ, the conservation of momentum implies the relation:

$$\boldsymbol{k}(\omega_1, \theta, \varphi) + \boldsymbol{k}(\omega_2, \theta, \varphi) - \boldsymbol{k}(\omega_3, \theta, \varphi) = \mathbf{O} \tag{3}$$

with

$$\boldsymbol{k}(\omega_i, \theta, \varphi) = [\omega_i/c]n(\omega_i, \theta, \varphi)\boldsymbol{u}(\theta, \varphi) \qquad i = 1, 2, 3 \tag{4}$$

ω_i is the circular frequency of wave i, c is the velocity of light in a vacuum and $n(\omega_i, \theta, \varphi)$ is the refractive index at ω_i in the direction of propagation with the unit vector $\boldsymbol{u}$ (θ, φ). θ and φ are the spherical coordinates relative to an orthonormal frame of which the axes X, Y, Z are the principal axes of the index ellipsoid. The different tensors studied will be expressed in this 'optical frame', linked to the crystallographic frame in accordance with the standard convention [4].

The cartesian coordinates (u_x, u_y, u_z) are related to the spherical coordinates (θ, φ) by the usual relations:

$$u_x = \cos\varphi \sin\theta \qquad u_y = \sin\varphi \sin\theta \qquad u_z = \cos\theta. \tag{5}$$

The three waves propagate in the same direction $\boldsymbol{u}$ (θ, φ). Therefore, according to (4), the phase-matching direction (3) is written:

$$\omega_1 n(\omega_1, \theta, \varphi) + \omega_2 n(\omega_2, \theta, \varphi) - \omega_3 n(\omega_3, \theta, \varphi) = 0. \tag{6}$$

2.1.2. The refractive indices in the direction $\boldsymbol{u}$ are given by the Fresnel equation which admits two solutions [5]:

$$n^+(\omega_i) = [2/(-B_i - (B_i^2 - 4C_i)^{1/2})]^{1/2} \tag{7}$$

and

$$n^-(\omega_i) = [2/(-B_i + (B_i^2 - 4C_i)^{1/2})]^{1/2} \qquad (n_+(\omega_i) > n_-(\omega_i)) \tag{8}$$

with

$$B_i = -u_x^2(b_i + c_i) - u_y^2(a_i + c_i) - u_z^2(a_i + b_i) \tag{9}$$

$$C_i = u_x^2 b_i c_i + u_y^2 a_i c_i + u_z^2 a_i b_i \tag{10}$$

with

$$a_i = n_x^{-2}(\omega_i) \qquad b_i = n_y^{-2}(\omega_i) \qquad c_i = n_z^{-2}(\omega_i) \tag{11}$$

$n_x(\omega_i)$, $n_y(\omega_i)$ and $n_z(\omega_i)$ are the principal refractive indices of the index ellipsoid at the circular frequency ω_i.

The two uniaxial and biaxial optical classes of positive and negative signs, are represented in figure 1 by their respective index surfaces from which the equations are given by relations (7) and (8).

2.1.3. The electric field vectors $\boldsymbol{E}^+$ and $\boldsymbol{E}^-$, associated with the indices n^+ and n^-, are calculated from the propagation equations projected on the three axes X, Y, Z. We obtain, for each wave, three equations which relate the three components (E_x, E_y, E_z) to the components (u_x, u_y, u_z) of the direction of propagation [6]:

$$\frac{E_p^+}{(n^+)^2} = \frac{u_p(u_x E_x^+ + u_y E_y^+ + u_z E_z^+)}{(n^+)^2 - (n_p)^2} \qquad \frac{E_p^-}{(n^-)^2} = \frac{u_p(u_x E_x^- + u_y E_y^- + u_z E_z^-)}{(n^-)^2 - (n_p)^2} \tag{12}$$

$p = x, y$ and z.

(i) In a negative uniaxial crystal, the polarization $\boldsymbol{E}^+$ is associated with the ordinary wave, symbolized by (o), which is contained in the (X, Y) plane. The polarization $\boldsymbol{E}^-$, orthogonal to $\boldsymbol{E}^+$, is associated with the extraordinary wave, (e) (see figure 1). The situation is inverted in a positive uniaxial crystal. The electric field components of the ordinary waves, $\boldsymbol{E}^{\mathrm{o}}$, and extraordinary waves, $\boldsymbol{E}^{\mathrm{e}}$, are the following:

$$\boldsymbol{E}^{\mathrm{o}} = E^{\mathrm{o}}\boldsymbol{e}^{\mathrm{o}} \qquad \text{with } \boldsymbol{e}^{\mathrm{o}} \text{ of components} \begin{pmatrix} -\sin\varphi \\ +\cos\varphi \\ 0 \end{pmatrix}$$

and (13)

$$\boldsymbol{E}^{\mathrm{e}} = E^{\mathrm{e}}\boldsymbol{e}^{\mathrm{e}} \qquad \text{with } \boldsymbol{e}^{\mathrm{e}} \text{ of components} \begin{pmatrix} -\cos[\theta + \rho(\theta)]\cos\varphi \\ -\cos[\theta + \rho(\theta)]\sin\varphi \\ +\sin[\theta + \rho(\theta)] \end{pmatrix}$$

with $\boldsymbol{E}^{\mathrm{e}} \cdot \boldsymbol{E}^{\mathrm{o}} = 0$.

$E^{\mathrm{o,e}}$ are the amplitudes of the ordinary and extraordinary waves and $\boldsymbol{e}^{\mathrm{o,e}}$ the associated unit vector. $\rho(\theta)$ is the birefringency angle given by:

$$\rho(\theta) = \arccos[\boldsymbol{D} \cdot \boldsymbol{E}]$$

where $\boldsymbol{D}$ is the electric displacement vector with

$$\boldsymbol{D} \cdot \boldsymbol{E} = (\cos^2\theta/n_{\mathrm{o}}^2 + \sin^2\theta/n_{\mathrm{e}}^2)(\cos^2\theta/n_0^4 + \sin^2\theta/n_{\mathrm{e}}^4)^{-1/2} \tag{14}$$

(ii) In a biaxial crystal, $\boldsymbol{E}^+$ and $\boldsymbol{E}^-$ are non-orthogonal except for the directions of propagation collinear to X, Y and Z. But the orthogonality of the electric displacement $\boldsymbol{D}^+$ and $\boldsymbol{D}^-$ is always verified. We shall keep the designation of 'ordinary' and 'extraordinary' waves and of 'positive' and 'negative' sign for the biaxial crystals. For a positive biaxial crystal, the ordinary wave has index n^+, only in the (X, Z) plane for the directions

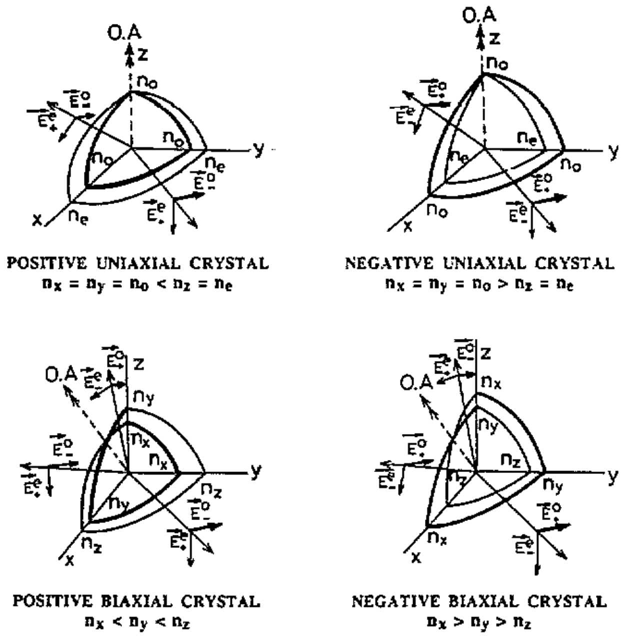


Figure 1. Index surfaces of the negative and positive uniaxial and biaxial optical classes. $E^{o,e}_{+,-}$ is the electric field of the ordinary or extraordinary wave associated with the external or internal index surface. O.A denotes the optical axis; the thick curves denote the ordinary wave and the fine curves the extraordinary wave.

of propagation contained between Z and the optical axis; the ordinary wave has index n^- everywhere else. The extraordinary wave has index n^- only between Z and the optical axis, and has index n^+ everywhere else. The situation is inverted for a negative biaxial crystal (figure 1). Thus, for a biaxial crystal, there is a discontinuity of the polarization on either side of the optical axis [7]. In other words, all biaxial crystals have the characteristic of a positive or negative uniaxial crystal according to the direction of propagation with respect to the optical axis. The ordinary wave only has index n_x and n_y in the principal plane of the index surfaces, that is the case in uniaxial crystals for each direction of propagation.

2.1.4. The dispersion in frequency of the refractive indices determines the number of possible combinations of index which verify the phase-matching relation.

According to (1) and (6), the relation between the refractive indices is:

$$[n(\omega_3) - n(\omega_2)]/[n(\omega_1) - n(\omega_3)] = \omega_1/\omega_2. \tag{15}$$

This ratio, necessarily positive, imposes:

$$n(\omega_1) < n(\omega_3) < n(\omega_2) \tag{16}$$

or

$$n(\omega_2) < n(\omega_3) < n(\omega_1). \tag{17}$$

Furthermore, the dispersion in frequency implies, according to (2):

$$n^+(\omega_3) > n^+(\omega_2) > n^+(\omega_1) \tag{18}$$

and

$$n^-(\omega_3) > n^-(\omega_2) > n^-(\omega_1). \tag{19}$$

According to (18) and (19), the relations (16) lead to the following phase-matching relations:

$$\omega_1 n^+(\omega_1) + \omega_2 n^+(\omega_2) - \omega_3 n^-(\omega_3) = 0 \tag{20}$$

and

$$\omega_1 n^-(\omega_1) + \omega_2 n^+(\omega_2) - \omega_3 n^-(\omega_3) = 0. \tag{21}$$

The inequalities (17) authorize just one phase-matching relation:

$$\omega_1 n^+(\omega_1) + \omega_2 n^-(\omega_2) - \omega_3 n^-(\omega_3) = 0. \tag{22}$$

The phase-matching conditions are related to the birefringence and to the dispersion in frequency of the refractive indices. In accordance with these parameters, Hobden [8] classifies the different types of cone generated by the phase-matching directions of the second harmonic generation (SHG).

For each phase-matching relation (20), (21) or (22) there is a specific configuration of polarization according to the 'optical sign' of the direction of propagation, which leads to six different configurations (table 1).

The type of phase-matching, I, II or III, is defined acording to the polarizations of the waves at ω_1 and ω_2: type I characterizes the interactions for which these two waves have the same polarization: (e)(e) or (o)(o). The two polarizations are different, (e)(o) or (o)(e), for the types II and III [9].

The type of phase-matching differs according to the interactions: SFM ($\omega_3 = \omega_1 + \omega_2$), DFM ($\omega_1 = \omega_3 - \omega_2$) and DFM ($\omega_2 = \omega_3 - \omega_1$) (table 1).

2.2. *Field factor and classification principle*

2.2.1. The non-linear interactions at three waves are governed by the second-order electric susceptibility tensor $\boldsymbol{\chi}^{(2)}$. This tensor of rank three has twenty-seven independent elements in the general case [3]:

$$\boldsymbol{\chi}^{(2)} = \begin{pmatrix} \chi_{XXX} & \chi_{XYY} & \chi_{XZZ} & \chi_{XYZ} & \chi_{XZY} & \chi_{XXZ} & \chi_{XZX} & \chi_{XXY} & \chi_{XYX} \\ \chi_{YXX} & \chi_{YYY} & \chi_{YZZ} & \chi_{YYZ} & \chi_{YZY} & \chi_{YXZ} & \chi_{YZX} & \chi_{YXY} & \chi_{YYX} \\ \chi_{ZXX} & \chi_{ZYY} & \chi_{ZZZ} & \chi_{ZYZ} & \chi_{ZZY} & \chi_{ZXZ} & \chi_{ZZX} & \chi_{ZXY} & \chi_{ZYX} \end{pmatrix} \tag{23}$$

X, Y and Z refer to the 'optical frame'.

The orientation symmetry [4] imposes relations between the components of the tensor $\boldsymbol{\chi}^{(2)}$ and so reduces the number of independent components. Furthermore there are other restrictions when the non-linear polarization is of electronic, rather than ionic, origin and when the crystal has a low absorption at the frequencies concerned (Kleinman's conditions) [10]. The nature and relative orientation of the anharmonic chemical bonds condition the magnitude and sign of the non-zero elements of the tensor [1, 2, 11].

2.2.2. The efficiency of the non-linear interaction between the three waves depends not only on $\boldsymbol{\chi}^{(2)}$ but on the configuration of polarization.

Table 1. Correspondence between the phase-matching relationships, the types of interaction, the polarization configuration and the optical sign of the propagation direction.

	Configuration of polarization						Type of phase-matching		
	Negative sign			Positive sign					
Phase-matching relations	ω_1	ω_2	ω_3	ω_1	ω_2	ω_3	SFM ($\omega_3 = \omega_1 + \omega_2$)	DFM ($\omega_1 = \omega_3 - \omega_2$)	DFM ($\omega_2 = \omega_3 - \omega_1$)
$\omega_1 n^+(\omega_1) + \omega_2 n^+ \omega_2 - \omega_3 n^-(\omega_3) = 0$	o	o	e	e	e	o	Type I	Type II	Type III
$\omega_1 n^-(\omega_1) + \omega_2 n^+(\omega_2) - \omega_3 n^-(\omega_3) = 0$	e	o	e	o	e	o	Type II	Type III	Type I
$\omega_1 n^+(\omega_1) + \omega_2 n^-(\omega_2) - \omega_3 n^-(\omega_3) = 0$	o	e	e	e	o	o	Type III	Type I	Type II

In the case of the SFM and for each direction of propagation with the coordinates (θ, φ), the effective coefficient, χ_{eff}, is expressed by [3]:

$$\chi_{eff}(\omega_3 = \omega_1 + \omega_2, \theta, \varphi) = e(\omega_3, \theta, \varphi)\chi^{(2)}(\omega_3) : e(\omega_1, \theta, \varphi)e(\omega_2, \theta, \varphi). \tag{24}$$

So:

$$\chi_{eff}(\omega_3 = \omega_1 + \omega_2, \theta, \varphi) = \sum_i e_i^*(\omega_3, \theta, \varphi) \sum_{jk} \chi_{ijk}(\omega_3) e_j(\omega_1, \theta, \varphi) e_k(\omega_2, \theta, \varphi). \tag{25}$$

For the DFM interactions, χ_{eff} is defined by:

$$\chi_{eff}(\omega_1 = \omega_3 - \omega_2, \theta, \varphi) = \sum_i e_i^*(\omega_1, \theta, \varphi) \sum_{jk} \chi_{ijk}(\omega_1) e_j(\omega_3, \theta, \varphi) e_k(\omega_2, \theta, \varphi) \tag{26}$$

$$\chi_{eff}(\omega_2 = \omega_3 - \omega_1, \theta, \varphi) = \sum_i e_i^*(\omega_2, \theta, \varphi) \sum_{jk} \chi_{ijk}(\omega_2) e_j(\omega_3, \theta, \varphi) e_k(\omega_1, \theta, \varphi). \tag{27}$$

The indices (i, j, k) refer to the 'optical frame' (X, Y, Z).

$e(\omega_p, \theta, \varphi)$ is the unit vector of the electric field of the wave with the circular frequency $\omega_p (p = 1, 2, 3)$, calculated from relations (12), following by normalization.

2.2.3. Earlier [12, 13], for the particular experimental study of the SHG and direct THG (third harmonic generation) in KTP, we have considered the product $e_i e_j e_k$ of the components of the electric fields as a single physical entity. We have called it the 'field factor' written F_{ijk} and defined by:

$$F_{ijk}(\omega_3 = \omega_1 + \omega_2, \theta, \varphi) = e_i^*(\omega_3, \theta, \varphi) e_j(\omega_1, \theta, \varphi) e_k(\omega_2, \theta, \varphi) \tag{28}$$

$$F_{ijk}(\omega_1 = \omega_3 - \omega_2, \theta, \varphi) = e_i^*(\omega_1, \theta, \varphi) e_j(\omega_3, \theta, \varphi) e_k(\omega_2, \theta, \varphi) \tag{29}$$

$$F_{ijk}(\omega_2 = \omega_3 - \omega_1, \theta, \varphi) = e_i^*(\omega_2, \theta, \varphi) e_j(\omega_3, \theta, \varphi) e_k(\omega_1, \theta, \varphi). \tag{30}$$

F_{ijk} corresponds to the contribution of χ_{ijk} to the effective coefficient χ_{eff}.

Therefore, the effective coefficient χ_{eff} of the SFM is a linear combination of the components χ_{ijk} and of the field factors F_{ijk}:

$$\chi_{eff}(\omega_3 = \omega_1 + \omega_2, \theta, \varphi) = \sum_{ijk} F_{ijk}(\omega_3 = \omega_1 + \omega_2, \theta, \varphi) \chi_{ijk}(\omega_3) \tag{31}$$

$$\chi_{eff}(\omega_1 = \omega_3 - \omega_2, \theta, \varphi) = \sum_{ijk} F_{ijk}(\omega_1 = \omega_3 - \omega_2, \theta, \varphi) \chi_{ijk}(\omega_1) \tag{32}$$

$$\chi_{eff}(\omega_2 = \omega_3 - \omega_1, \theta, \varphi) = \sum_{ijk} F_{ijk}(\omega_2 = \omega_3 - \omega_1, \theta, \varphi) \chi_{ijk}(\omega_2). \tag{33}$$

For the SFM, the first tensor index is relative to the wave at ω_3, the second to the wave at ω_1 and the third to the wave at ω_2. For the DFM ($\omega_1 = \omega_3 - \omega_2$), the first, second and third tensor indices are relative to the waves at ω_1, ω_3 and ω_2 respectively and for the DFM ($\omega_2 = \omega_3 - \omega_1$) to the waves at ω_2, ω_3 and ω_1 respectively.

Thus the field factor is a tensor of rank three which has twenty-seven independent elements in the general case:

$$\mathbf{F}=\begin{pmatrix} F_{XXX} & F_{XYY} & F_{XZZ} & F_{XYZ} & F_{XZY} & F_{XXZ} & F_{XZX} & F_{XXY} & F_{XYX} \\ F_{YXX} & F_{YYY} & F_{YZZ} & F_{YYZ} & F_{YZY} & F_{YXZ} & F_{YZX} & F_{YXY} & F_{YYX} \\ F_{ZXX} & F_{ZYY} & F_{ZZZ} & F_{ZYZ} & F_{ZZY} & F_{ZXZ} & F_{ZZX} & F_{ZXY} & F_{ZYX} \end{pmatrix}. \qquad (34)$$

Each component F_{ijk} is a trigonometric function which depends on the direction of propagation, in this case the phase-matching direction, so depends only on the linear optical properties of crystal.

The optical class, uniaxial and biaxial, and the properties of orthogonality and collinearity of the coupled electric fields impose restrictions and relations between elements and so reduce the number of independent components, an aspect we hope to study subsequently.

According to the configuration of polarization, each family comes in the form of three different tensors, written (eoo), (ooe), (eoe) for the family 2o.e and (oee), (eeo), (eoe) for the family 2e.o. The correspondence between polarizations and frequencies is given in table 2 in the eighteen possible cases. It is not an arbitrary re-assembly: it is based on the properties of orthogonality and collinearity of the electric fields of the three waves.

Table 2. Classification in two families, 2o.e and 2e.o, of the eighteen possible cases for the 3-wave non-linear optical interactions. Correspondence between the types of interaction, the optical signs of propagation directions, the polarization configurations, the circular frequencies and the tensor indices.

Type 2o.e Configuration of polarization					Type 2e.o Configuration of polarization				
SFM					SFM				
$\omega_3=\omega_1+\omega_2$		ω_3	ω_1	ω_2	$\omega_3=\omega_1+\omega_2$		ω_3	ω_1	ω_2
Type I		e	o	o	Type I		o	e	e
<0	Réf.	(i)	(j)	(k)	>0	Réf.	(i)	(j)	(k)
Type II		o	o	e	Type II		e	e	o
>0		(k)	(j)	(i)	<0		(k)	(j)	(i)
Type III		o	e	o	Type III		e	o	e
>0		(k)	(i)	(j)	<0		(k)	(i)	(j)
DFM					DFM				
$\omega_1=\omega_3-\omega_2$		ω_1	ω_3	ω_2	$\omega_1=\omega_3-\omega_2$		ω_1	ω_3	ω_2
Type I		e	o	o	Type I		o	e	e
>0		(i)	(k)	(j)	<0		(i)	(k)	(j)
Type II		o	e	o	Type II		e	o	e
<0		(j)	(i)	(k)	>0		(j)	(i)	(k)
Type III		o	o	e	Type III		e	e	o
>0		(j)	(k)	(i)	<0		(j)	(k)	(i)
DFM					DFM				
$\omega_2=\omega_3-\omega_1$		ω_2	ω_3	ω_1	$\omega_2=\omega_3-\omega_1$		ω_2	ω_3	ω_1
Type I		e	o	o	Type I		o	e	e
>0		(i)	(k)	(j)	<0		(i)	(k)	(j)
Type II		o	o	e	Type II		e	e	o
>0		(j)	(k)	(i)	<0		(j)	(k)	(i)
Type III		o	e	o	Type III		e	o	e
<0		(k)	(i)	(j)	>0		(k)	(i)	(j)

(i) In uniaxial crystals, the electric fields of the ordinary and extraordinary waves verify the following relation:

$$e^{o}(\omega_i) \cdot e^{e}(\omega_j) = 0 \qquad \text{with } i, j = 1, 2 \text{ or } 3. \tag{35}$$

The orthogonality of the electric fields of the waves corresponding to the two first suffices, to the two last suffices and to the first and third suffices leads respectively to the three relations:

$$F_{xxi} + F_{yyi} + F_{zzi} = 0 \tag{36}$$

$$F_{ixx} + F_{iyy} + F_{izz} = 0 \tag{37}$$

$$F_{xix} + F_{yiy} + F_{ziz} = 0 \tag{38}$$

where $i = x, y, z$.

Hence, in uniaxial crystals, the elements of tensors of types (eoo) and (oee) obey the equalities (36), (38), those of types (ooe) and (eeo) obey (37), (38) and those of types (oeo) and (eoe) obey the equalities (36) and (37).

(ii) In biaxial crystals, the relations (36), (37) and (38) are verified only for the directions of propagation collinear to the principal axes of the index surface. For other directions, these equalities are not strict, especially since n_x differs from n_y.

2.2.4. The field factor is suited to the study of the relationships between SFM and DFM, which leads to the classification of all these interactions:

(i) For a given interaction, the permutation of the polarizations (extraordinary and ordinary) without the permutation of the associated frequencies, allows the interactions of types I, II and III for the directions of propagation with different optical signs to be grouped together. There are two families, each grouping together nine situations: the family, written 2o.e, in which two ordinary waves are coupled with one extraordinary wave, and the family 2e.o, for which two extraordinary waves are coupled with one ordinary wave. The permutations of the tensor indices (i, j, k) associated with the permutation of the polarizations are given in table 2. We take the SFM of type I in the negative class for the family 2o.e and the SFM of type I in the positive class for the family 2e.o as references for these permutations.

(ii) Furthermore, for a class of a given sign, the permutation of frequencies with the permutation of polarizations governs the relationships between SFM and DFM. For each phase-matching direction the equalities are:

$$\begin{aligned}
F^{\mathrm{I}}_{ijk}(\omega_3 = \omega_1 + \omega_2) &= F^{\mathrm{II}}_{jik}(\omega_1 = \omega_3 - \omega_2) = F^{\mathrm{III}}_{kij}(\omega_2 = \omega_3 - \omega_1)\\
F^{\mathrm{II}}_{ijk}(\omega_3 = \omega_1 + \omega_2) &= F^{\mathrm{III}}_{jik}(\omega_1 = \omega_3 - \omega_2) = F^{\mathrm{I}}_{kij}(\omega_2 = \omega_3 - \omega_1)\\
F^{\mathrm{III}}_{ijk}(\omega_3 = \omega_1 + \omega_2) &= F^{\mathrm{I}}_{jik}(\omega_1 = \omega_3 - \omega_2) = F^{\mathrm{II}}_{kij}(\omega_2 = \omega_3 - \omega_1).
\end{aligned} \tag{39}$$

3. Uniaxial classes

The typical field factors of the two interaction families are calculated for the SFM ($\lambda_1 = 1.32\ \mu\mathrm{m}, \lambda_2 = 0.66\ \mu\mathrm{m}, \lambda_3 = 0.44\ \mu\mathrm{m}$) in type I phase-matching condition in two typical uniaxial crystals of which the dispersions in frequency of the refractive indices and of the birefringence are comparable to those of usual non-linear crystals at optical frequencies: one with positive sign for the study of the family 2e.o and the other with a negative sign for the family 2o.e. The associated phase-matching directions are cal-

UNIAXIAL CRYSTAL

	POSITIVE		NEGATIVE	
λ (μm)	n_o	n_e	n_o	n_e
1,32	1,7352	1,8212	1,8212	1,7352
0,66	1,7642	1,8611	1,8611	1,7642
0,44	1,8152	1,9360	1,9360	1,8152

BIAXIAL CRYSTAL

	POSITIVE CONE TYPE A			NEGATIVE CONE TYPE A		
λ (μm)	n_x	n_y	n_z	n_x	n_y	n_z
1	1,7550	1,7600	1,8250	1,8400	1,8200	1,7550
$\frac{1}{2}$	1,8050	1,8100	1,8750	1,8851	1,8680	1,8050
$\frac{1}{3}$	1,8300	1,8500	1,9100	1,9100	1,9060	1,8450

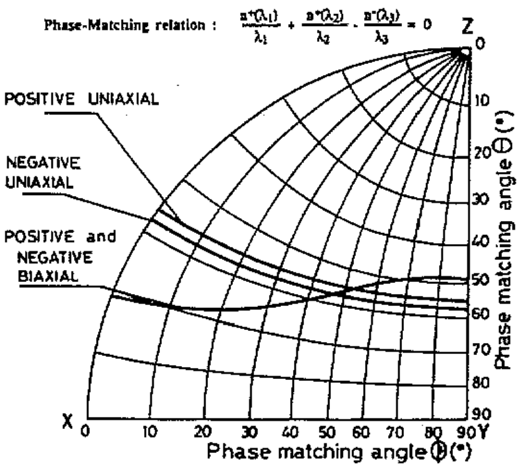


Figure 2. Refractive indices and SFM type I phase-matching directions of uniaxial and biaxial crystals for the field factors calculation. The phase-matching relation is $n^+(\lambda_1)/\lambda_1 + n^+(\lambda_2)/(\lambda_2) - n^-(\lambda_3)/\lambda_3 = 0$.

culated from (20) with the refractive indices given in figure 2. The spherical coordinates (θ, φ) of the phase-matching directions are drawn on the Wülf diagram of figure 2. The two crystals differ only by the inversion of the values of the ordinary and extraordinary indices which leads to two adjacent phase-matching cones and thus to field factors with comparable magnitudes.

3.1. *Field factors of interactions with a polarization configuration of type 2o.e*

Twelve field factors are non-zero for the type 2o.e and are described by twelve trigonometric functions with the following θ and φ variations:

$$
\begin{aligned}
&(1) = \sin[\theta + \rho(\omega_q, \theta)] \sin^2\varphi \qquad (\bar{1}) = \sin[\theta + \rho(\omega_q, \theta)] \cos^2\varphi \\
&(2) = -\cos[\theta + \rho(\omega_q, \theta)] \cos^3\varphi \qquad (\bar{2}) = -\cos[\theta + \rho(\omega_q, \theta)] \sin^3\varphi \\
&(3a) = (3b) = -\sin[\theta + \rho(\omega_q, \theta)] \sin\varphi \cos\varphi \\
&(4a) = (4b) = -(5) = \cos[\theta + \rho(\omega_q, \theta)] \cos^2\varphi \sin\varphi \\
&(\bar{4}a) = (\bar{4}b) = -(\bar{5}) = \cos[\theta + \rho(\omega_q, \theta)] \cos\varphi \sin^2\varphi .
\end{aligned}
\tag{40}
$$

The notations such as (1) and ($\bar{1}$) indicate that the two functions are out of phase by $\pi/2$ in φ. This notation bears no relation to the written convention of Hermann–Maugin for the orientation classes of symmetry. For a given phase-matching direction cone, θ is constant for all values of φ. The birefringence angle $\rho(\omega_q, \theta)$ is given by relation (14).

The φ angular variation of each function is the same for all the 2o.e interactions. But the θ angular variation depends on the considered interaction according to the following relations (39).

$\omega_q = \omega_1$ for:

SFM($\omega_3 = \omega_1 + \omega_2$) type III DFM($\omega_1 = \omega_3 - \omega_2$) type I

DFM($\omega_2 = \omega_3 - \omega_1$) type II.

$\omega_q = \omega_2$ for:

SFM($\omega_3 = \omega_1 + \omega_2$) type II DFM($\omega_2 = \omega_3 - \omega_1$) type I

DFM($\omega_1 = \omega_3 - \omega_2$) type III.

$\omega_q = \omega_3$ for:

SFM($\omega_3 = \omega_1 + \omega_2$) type I DFM($\omega_1 = \omega_3 - \omega_2$) type II

DFM($\omega_2 = \omega_3 - \omega_1$) type III.

$\rho(\omega_1)$, $\rho(\omega_2)$ and $\rho(\omega_3)$ are different in accordance with the dispersion in frequency of the birefringence. These functions are drawn in figure 3 for values of φ contained between 0 and $\pi/2$, φ being the spherical coordinate of each type I phase-matching direction for the SFM in the negative typical crystal.

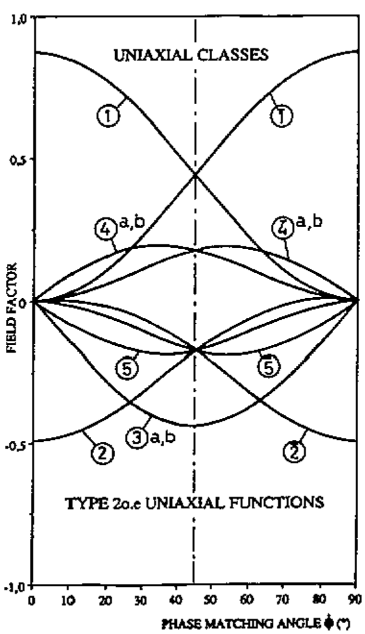


Figure 3. φ angular variation of type 2o.e field factors in uniaxial crystals.

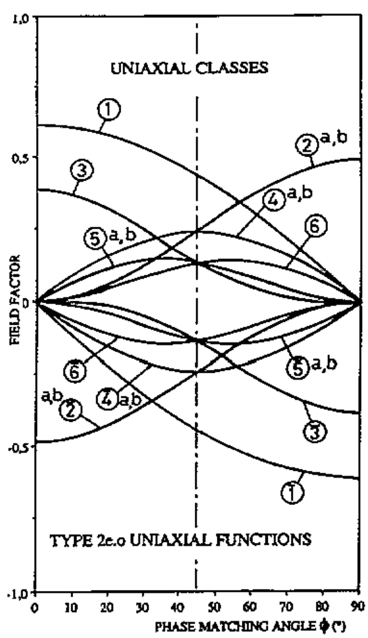


Figure 4. φ angular variation of type 2e.o field factors in uniaxial crystals.

The calculation of the twelve field factors, for each of the eight other cases of the family 2o.e, leads to the same set of nine trigonometric functions. But to each function corresponds a tensor element characteristic of a given case. The relation between the trigonometric functions and the elements of the field factor tensor are given in table 3. The equalities (39) relate SFM and DFM of types I, II and III.

The three tensors (eoo), (ooe) and (oeo) are described in table 4. They have twelve non-zero elements of which four are independent.

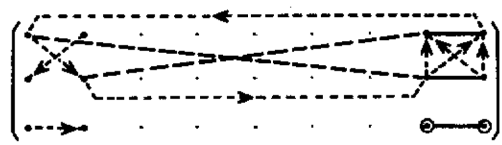
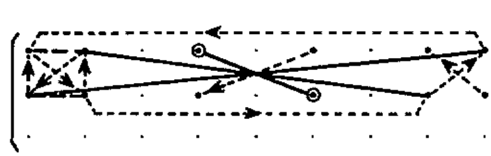
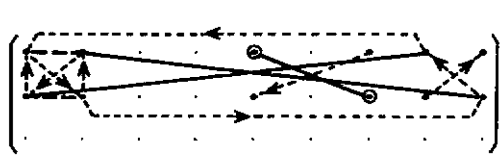
Thus, for a crystal belonging to a given class of orientation symmetry, the elements of the second order electric susceptibility tensor are not all involved, and vary in accordance with the configuration of polarization of the beam: (eoo), (ooe) or (oeo). Conversely, for a given configuration of polarization, the elements of the field factor tensor are not all involved and vary according to the orientation symmetry of the crystal.

Tables 5(a) and 5(b) collect the characteristic functions of the non-linear interaction of the family 2o.e for the thirteen non-centrosymmetric uniaxial classes with the Scho-

Table 3. Correspondence between the type 2o.e uniaxial functions and the F_{ijk} coefficients associated with the different interactions of the family 2o.e.

<table>
<tr><th rowspan="2">Interactions</th><th rowspan="2">Optical sign</th><th colspan="9">Type 2o.e or 2o.e 2e.o Uniaxial functions</th></tr>
<tr><th>1</th><th>$\bar{1}$</th><th>2</th><th>$\bar{2}$</th><th>3
3a 3b</th><th>4
4a 4b</th><th>$\bar{4}$
$\bar{4}$a $\bar{4}$b</th><th>5</th><th>$\bar{5}$</th></tr>
<tr><td>SFM
$\omega_3 = \omega_1 + \omega_2$
Type I</td><td><0 Uniaxial
<0 Biaxial</td><td>F_{ZYY}</td><td>F_{ZXX}</td><td>F_{XYY}</td><td>F_{YXX}</td><td>=
≅
F_{ZXY}, F_{ZYX}</td><td>=
≅
F_{XXY}, F_{XYX}</td><td>=
≅
F_{YXY}, F_{YYX}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
<tr><td>SFM
$\omega_3 = \omega_1 + \omega_2$
Type II</td><td>>0 Uniaxial
>0 Biaxial</td><td>F_{YYZ}</td><td>F_{XXZ}</td><td>F_{YYX}</td><td>F_{XXY}</td><td>=
≅
F_{YXZ}, F_{XYZ}</td><td>=
≅
F_{YXX}, F_{XYX}</td><td>=
≅
F_{YXY}, F_{XYY}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
<tr><td>SFM
$\omega_3 = \omega_1 + \omega_2$
Type III</td><td>>0 Uniaxial
>0 Biaxial</td><td>F_{YZY}</td><td>F_{XZX}</td><td>F_{YXY}</td><td>F_{XYX}</td><td>=
≅
F_{YZX}, F_{XZY}</td><td>=
≅
F_{YXX}, F_{XXY}</td><td>=
≅
F_{YYX}, F_{XYY}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
<tr><td>DFM
$\omega_2 = \omega_3 - \omega_1$
Type III</td><td><0 Uniaxial
<0 Biaxial</td><td>F_{YZY}</td><td>F_{XZX}</td><td>F_{YXY}</td><td>F_{XYX}</td><td>=
≅
F_{YZX}, F_{XZY}</td><td>=
≅
F_{YXX}, F_{XXY}</td><td>=
≅
F_{YYX}, F_{XYY}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
<tr><td>DFM
$\omega_1 = \omega_3 - \omega_2$
Type I
DFM
$\omega_2 = \omega_3 - \omega_1$
Type I</td><td>>0 Uniaxial
>0 Biaxial</td><td>F_{ZYY}</td><td>F_{ZXX}</td><td>F_{XYY}</td><td>F_{YXX}</td><td>=
≅
F_{ZYX}, F_{ZXY}</td><td>=
≅
F_{XYX}, F_{XXY}</td><td>=
≅
F_{YYX}, F_{YXY}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
<tr><td>DFM
$\omega_1 = \omega_3 - \omega_2$
Type II</td><td><0 Uniaxial
<0 Biaxial</td><td>F_{YZY}</td><td>F_{XZX}</td><td>F_{YXY}</td><td>F_{XYX}</td><td>=
≅
F_{XZY}, F_{YZX}</td><td>=
≅
F_{XXY}, F_{YXX}</td><td>=
≅
F_{XYY}, F_{YYX}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
<tr><td>DFM
$\omega_1 = \omega_3 - \omega_2$
Type III
DFM
$\omega_2 = \omega_3 - \omega_1$
Type II</td><td>>0 Uniaxial
>0 Biaxial</td><td>F_{YYZ}</td><td>F_{XXZ}</td><td>F_{YYX}</td><td>F_{XXY}</td><td>=
≅
F_{XYZ}, F_{YXZ}</td><td>=
≅
F_{XYX}, F_{YXX}</td><td>=
≅
F_{XYY}, F_{YXY}</td><td>F_{YYY}</td><td>F_{XXX}</td></tr>
</table>

Table 4. Type (eoo), (ooe) and (oeo) field factor tensors. · $F_{ijk}(\varphi)=0$; ● $F_{ijk}(\varphi)\neq 0$, periodicity π; ⊙ $F_{ijk}(\varphi)\neq 0$, periodicity $\pi/2$. —— $F_{ijk}(\varphi)=F_{lmn}(\varphi)$; - - - - $F_{ijk}(\varphi)=-F_{lmn}(\varphi)$; - - - $F_{ijk}(\varphi)=F_{lmn}(\varphi+\pi/2)$.

Interaction		Type 2o.e F_{ijk} field factor tensor for uniaxial classes
	Type eoo	XX YY ZZ YZ ZY XZ ZX XY YX
SFM type I < 0 $\omega_3=\omega_1+\omega_2$ DFM type I > 0 $\omega_1=\omega_3-\omega_2$ $\omega_2=\omega_3-\omega_1$	X Y Z	
	Type ooe	XX YY ZZ YZ ZY XZ ZX XY YX
SFM type II > 0 $\omega_3=\omega_1+\omega_2$ DFM type III > 0 $\omega_1=\omega_3-\omega_2$ DFM type II > 0 $\omega_2=\omega_3-\omega_1$	X Y Z	
	Type oeo	XX YY ZZ YZ ZY XZ ZX XY YX
SFM type III > 0 $\omega_3=\omega_1+\omega_2$ DFM type II < 0 $\omega_1=\omega_3-\omega_2$ DFM type III < 0 $\omega_2=\omega_3-\omega_1$	X Y Z	

enflies written convention. The corresponding tensor elements are read in table 3. The same elements F_{ijk} are involved in classes C_4 and C_6. It is the same for the classes $4mm$ and $6mm$, as for the classes D_4 and D_6.

For the classes of symmetry C_3, D_3, C_4, C_6, D_4 and D_6, the involvement of the function (3), relative to the field factors with X, Y and Z indices, depends on the considered interaction as table 5(b) shows. Indeed, the χ_{zxy} and χ_{zyx} elements of the electric susceptibility tensor in these six classes are zero whereas χ_{xyz}, χ_{xzy}, χ_{yxz} and χ_{yzx} are non-zero.

The effective coefficient of all interactions with a type 2o.e configuration of polarization is nil in the crystals belonging to the classes of symmetry D_4 and D_6. These two classes have the same $\chi^{(2)}$ tensor and the only non-zero components are $\chi_{xyz}=-\chi_{yxz}$ and $\chi_{xzy}=-\chi_{yzx}$. Thus, according to the field factor tensors of table 4, the effective coefficient is nil for all phase-matching directions.

For the types eoo, ooe and oeo, respectively:

$$\chi_{\text{eff}}=0(\chi_{xyz}+\chi_{yxz})+0(\chi_{xzy}+\chi_{yzx})=0. \tag{41}$$

$$\chi_{\text{eff}}=F_{xyz}(\chi_{xyz}+\chi_{yxz})+0(\chi_{xzy}+\chi_{yzx})=0. \tag{42}$$

$$\chi_{\text{eff}}=0(\chi_{xyz}+\chi_{yxz})+F_{zxy}(\chi_{xzy}+\chi_{yzx})=0. \tag{43}$$

Table 5(a). Intervening 2o.e and 2e.o uniaxial functions for the thirteen non-centrosymmetric uniaxial classes of symmetry. Asterisks denote functions for which the involvement depends on interaction. The correspondence is given in table 5(b). The standard frame of orientation (x_1, x_2, x_3) is collinear with the 'optical frame' (X, Y, Z).

Uniaxial acentric classes of symmetry	C_3	C_{3v} $(m \perp x_1)$	C_{3v} $(m \perp x_2)$	C_{3h}	D_3	D_{3h} $(m \perp x_1)$	D_{3h} $(m \perp x_2)$	S_4	C_4, C_6	C_{4v} C_{6v}	D_{2d} $(m \parallel x_1)$	D_4 D_6
Intervening type 2o.e or 2o.e 2e.o functions	1, $\bar{1}$, 2, $\bar{2}$, 3*, 4a, 4b $\bar{4}$a, $\bar{4}$b, 5, $\bar{5}$	1, $\bar{1}$, $\bar{2}$, 4a, 4b, 5	1, $\bar{1}$, 2, $\bar{4}$a, $\bar{4}$b, $\bar{5}$	2, $\bar{2}$, $\bar{4}$a, $\bar{4}$b, 5, $\bar{5}$	2, 3*, $\bar{4}$a, $\bar{4}$b, $\bar{5}$	$\bar{2}$, 4a, 4b, 5	2, $\bar{4}$a, $\bar{4}$b. $\bar{5}$	1, $\bar{1}$, 3a, 3b	1, $\bar{1}$, 3*	1, $\bar{1}$	3a, 3b	3*
Intervening type 2e.o or 2e.o 2o.e functions	2*, $\bar{2}$*, 3, $\bar{3}$, 4a, 4b, $\bar{4}$a, $\bar{4}$b, 5a, 5b, $\bar{5}$a, $\bar{5}$b, 6, $\bar{6}$	3, 4a, 4b, $\bar{4}$a, $\bar{4}$b, $\bar{5}$a, $\bar{5}$b, 6	$\bar{3}$, 4a, 4b, $\bar{4}$a, $\bar{4}$b, 5a, 5b, $\bar{6}$	3, $\bar{3}$, 5a, 5b, 6, $\bar{6}$	2*, $\bar{2}$*, $\bar{3}$, 5a, 5b, $\bar{6}$	3, $\bar{5}$a, $\bar{5}$b, 6	$\bar{3}$, 5a, 5b, $\bar{6}$	2a, 2b, $\bar{2}$a, $\bar{2}$b, 4a, 4b, $\bar{4}$a, $\bar{4}$b	2*, $\bar{2}$*, 4a, 4b, $\bar{4}$a, $\bar{4}$b	4a, 4b $\bar{4}$a, $\bar{4}$b	2a, 2b, $\bar{2}$a, $\bar{2}$b	2*, $\bar{2}$*

Table 5(b). Involvement of the functions (3a) and (3b) of type 2o.e and of the functions (2a), (2b), ($\bar{2}$a) and ($\bar{2}$b) of type 2e.o, according to the different interactions, for the uniaxial classes of symmetry C_3, D_3, C_4, C_6, D_4 and D_6. Once again, asterisks denote functions for which the involvement depends on interaction.

Interactions	Type 2o.e 3*	Type 2e.o 2*	Type 2e.o $\bar{2}$*
SFM ($\omega_3 = \omega_1 + \omega_2$) Type I	any	2a, 2b	$\bar{2}$a, $\bar{2}$b
SFM ($\omega_3 = \omega_1 + \omega_2$) Type II	3a, 3b	2b	$\bar{2}$b
SFM ($\omega_3 = \omega_1 + \omega_2$) Type III	3a, 3b	2b	$\bar{2}$b
DFM ($\omega_2 = \omega_3 - \omega_1$) Type III	3a, 3b	2b	$\bar{2}$b
DFM ($\omega_1 = \omega_3 - \omega_2$) DFM ($\omega_2 = \omega_3 - \omega_1$) Type I	any	2a, 2b	$\bar{2}$a, $\bar{2}$b
DFM ($\omega_1 = \omega_3 - \omega_2$) Type II	3a, 3b	2a	$\bar{2}$a
DFM ($\omega_1 = \omega_3 - \omega_2$) Type III DFM ($\omega_2 = \omega_3 - \omega_1$) Type II	3a, 3b	2a	$\bar{2}$a

3.2. *Field factors of interactions with a polarization configuration of type 2e.o*

The interactions of type 2e.o have eighteen non-zero field factors described by eighteen different trigonometric functions with the following angular variations:

$$
\begin{aligned}
(1) &= \cos\varphi \sin[\theta + \rho(\omega_q, \theta)] \sin[\theta + \rho(\omega_r, \theta)] \\
(\bar{1}) &= -\sin\varphi \sin[\theta + \rho(\omega_q, \theta)] \sin[\theta + \rho(\omega_r, \theta)] \\
(2\mathrm{a}) &= \sin^2\varphi \cos[\theta + \rho(\omega_q, \theta)] \sin[\theta + \rho(\omega_r, \theta)] \\
(2\mathrm{b}) &= \sin^2\varphi \sin[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)] \\
(\bar{2}\mathrm{a}) &= \cos^2\varphi \cos[\theta + \rho(\omega_q, \theta)] \sin[\theta + \rho(\omega_r, \theta)] \\
(\bar{2}\mathrm{b}) &= \cos^2\varphi \sin[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)] \\
(3) &= \cos^3\varphi \cos[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)] \\
(\bar{3}) &= -\sin^3\varphi \cos[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)] \\
(4\mathrm{a}) &= -(\bar{4}\mathrm{a}) = \sin\varphi \cos\varphi \cos[\theta + \rho(\omega_q, \theta)] \sin[\theta + \rho(\omega_r, \theta)] \\
(4\mathrm{b}) &= -(\bar{4}\mathrm{b}) = \sin\varphi \cos\varphi \sin[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)] \\
(5\mathrm{a}) &= (5\mathrm{b}) = -(6) = \sin\varphi \cos^2\varphi \cos[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)] \\
(\bar{5}\mathrm{a}) &= (\bar{5}\mathrm{b}) = -(\bar{6}) = -\sin^2\varphi \cos\varphi \cos[\theta + \rho(\omega_q, \theta)] \cos[\theta + \rho(\omega_r, \theta)].
\end{aligned}
\tag{44}
$$

As for the type 2o.e, the φ angular variation of each function is the same for all the 2e.o interactions. The θ angular variation depends on the dispersion in frequency of the birefringence angle ρ. The frequencies ω_q and ω_r of relations (44) are the following.

$(\omega_q, \omega_r) = (\omega_1, \omega_2)$ for:

SFM($\omega_3 = \omega_1 + \omega_2$) type I DFM($\omega_1 = \omega_3 - \omega_2$) type II

DFM($\omega_2 = \omega_3 - \omega_1$) type III.

$(\omega_q, \omega_r) = (\omega_3, \omega_1)$ for:

SFM($\omega_3 = \omega_1 + \omega_2$) type II DFM($\omega_1 = \omega_3 - \omega_2$) type III

DFM($\omega_2 = \omega_3 - \omega_1$) type I.

$(\omega_q, \omega_r) = (\omega_3, \omega_2)$ for:

SFM($\omega_3 = \omega_1 + \omega_2$) type III DFM($\omega_1 = \omega_3 - \omega_2$) type I

DFM($\omega_2 = \omega_3 - \omega_1$) type II.

The functions (2a) and (2b) have the same φ variations but differ in θ. It is the same for the functions (4a) and (4b) and so for the functions ($\bar{4}$a) and ($\bar{4}$b). The functions (5a) and (5b) are equal. It is the same for ($\bar{5}$a) and ($\bar{5}$b). Thus, the sixteen functions can be grouped into twelve functions with different φ equations: (1); ($\bar{1}$); (2) for the functions (2a, 2b); ($\bar{2}$) for ($\bar{2}$a, $\bar{2}$b); (3); ($\bar{3}$); (4) for (4a, 4b); ($\bar{4}$) for ($\bar{4}$a, $\bar{4}$b); (5) for (5a, 5b); ($\bar{5}$) for ($\bar{5}$a, $\bar{5}$b); (6) and ($\bar{6}$).

The functions of the couples (1, $\bar{1}$), (2, $\bar{2}$), (3, $\bar{3}$), (4, $\bar{4}$), (5, $\bar{5}$) and (6, $\bar{6}$) are out of phase by $\pi/2$ in φ.

The functions are drawn in figure 4, in the special case of the SFM of type I, phase-matched in the positive typical crystal for which the refractive indices are given in figure 2. The associated field factors are given in table 6.

The functions (2a) and (2b) are joined because the dispersion in frequency of the birefringence angle is small in this particular case, this is usually true. It is the same for the functions ($\bar{2}$a, $\bar{2}$b), (4a, 4b) and ($\bar{4}$a, $\bar{4}$b).

The three tensors for the configurations of polarization (oee), (eeo) and (eoe) are described in table 7. The tensors have eighteen non-zero elements for which seven are strictly independent. The type 2e.o functions involved are given in tables 5(a) and 5(b) for the thirteen non-centrosymmetric uniaxial classes. The re-assembly of the classes of symmetry for the involvement of the type 2e.o functions is the same as for the type 2o.e. The specific case of the field factors of the X, Y and Z indices concerns the functions (2a), (2b), ($\bar{2}$a) and ($\bar{2}$b).

The functions (1) and ($\bar{1}$) are never involved because the elements χ_{yzz}, χ_{zyz}, χ_{zzy}, χ_{xzz}, χ_{zxz} and χ_{zzx} are zero for all the non-centrosymmetric uniaxial classes of symmetry.

The classes C_{4v} and C_{6v} forbid all interaction of type 2e.o for similar reasons to the classes D_4 and D_6 with the interactions of type 2o.e.

4. Biaxial classes

The electric field vectors of the field factors must be calculated from the general relations (12). Indeed, the relations (13), (14), (40) and (44) are only valid in the principal planes of biaxial crystals and for all directions of propagation in uniaxial crystals. But we keep

Table 6. Correspondence between the type 2e.o uniaxial functions and the F_{ijk} coefficients relative to the different interactions of the family 2e.o.

		Type 2e.o or 2e.o 2o.e uniaxial functions											
Interactions	Optical sign	1	$\bar{1}$	2 2a 2b	$\bar{2}$ $\bar{2}$a $\bar{2}$b	3	$\bar{3}$	4 4a 4b	$\bar{4}$ $\bar{4}$a $\bar{4}$b	5 5a 5b	$\bar{5}$ $\bar{5}$a $\bar{5}$b	6	$\bar{6}$
SFM $\omega_3 = \omega_1 + \omega_2$ Type I	>0 Uniaxial >0 Biaxial	F_{YZZ}	F_{XZZ}	≅ ≅ F_{XYZ}, F_{XZY}	≅ ≅ F_{YXZ}, F_{YZX}	F_{YXX}	F_{XYY}	≅ ≅ F_{XXZ}, F_{XZX}	≅ ≅ F_{YYZ}, F_{YZY}	= ≅ F_{YXY}, F_{YYX}	= ≅ F_{XYX}, F_{XXY}	F_{YYY}	F_{XXX}
SFM $\omega_3 = \omega_1 + \omega_2$ Type II	<0 Uniaxial <0 Biaxial	F_{ZZY}	F_{ZZX}	≅ ≅ F_{ZYX}, F_{YZX}	≅ ≅ F_{ZXY}, F_{XZY}	F_{XXY}	F_{YYX}	≅ ≅ F_{ZXX}, F_{XZX}	≅ ≅ F_{ZYY}, F_{YZY}	= ≅ F_{YXY}, F_{XYY}	= ≅ F_{XYX}, F_{YXX}	F_{YYY}	F_{XXX}
SFM $\omega_3 = \omega_1 + \omega_2$ Type III	<0 Uniaxial <0 Biaxial	F_{ZYZ}	F_{ZXZ}	≅ ≅ F_{ZXY}, F_{YXZ}	≅ ≅ F_{ZYX}, F_{XYZ}	F_{XYX}	F_{YXY}	≅ ≅ F_{ZXX}, F_{XXZ}	≅ ≅ F_{ZYY}, F_{YYZ}	= ≅ F_{YYX}, F_{XYY}	= ≅ F_{XXY}, F_{YXX}	F_{YYY}	F_{XXX}
DFM $\omega_2 = \omega_3 - \omega_1$ Type III	>0 Uniaxial >0 Biaxial	F_{ZYZ}	F_{ZXZ}	≅ ≅ F_{ZXY}, F_{YXZ}	≅ ≅ F_{ZYX}, F_{XYZ}	F_{XYX}	F_{YXY}	≅ ≅ F_{ZXX}, F_{XXZ}	≅ ≅ F_{ZYY}, F_{YYZ}	= ≅ F_{YYX}, F_{XYY}	= ≅ F_{XXY}, F_{YXX}	F_{YYY}	F_{XXX}
DFM $\omega_1 = \omega_3 - \omega_2$ Type I DFM $\omega_2 = \omega_3 - \omega_1$ Type I	<0 Uniaxial <0 Biaxial	F_{YZZ}	F_{XZZ}	≅ ≅ F_{XZY}, F_{XYZ}	≅ ≅ F_{YZX}, F_{YXZ}	F_{YXX}	F_{XYY}	≅ ≅ F_{XZX}, F_{XXZ}	≅ ≅ F_{YZY}, F_{YYZ}	= ≅ F_{YYX}, F_{YXY}	= ≅ F_{XXY}, F_{XYX}	F_{YYY}	F_{XXX}
DFM $\omega_1 = \omega_3 - \omega_2$ Type II	>0 Uniaxial >0 Biaxial	F_{ZYZ}	F_{ZXZ}	≅ ≅ F_{YXZ}, F_{ZXY}	≅ ≅ F_{XYZ}, F_{ZYX}	F_{XYX}	F_{YXY}	≅ ≅ F_{XXZ}, F_{ZXX}	≅ ≅ F_{YYZ}, F_{ZYY}	= ≅ F_{XYY}, F_{YYX}	= ≅ F_{YXX}, F_{XXY}	F_{YYY}	F_{XXX}
DFM $\omega_1 = \omega_3 - \omega_2$ Type III DFM $\omega_2 = \omega_3 - \omega_1$ Type II	<0 Uniaxial <0 Biaxial	F_{ZZY}	F_{ZZX}	≅ ≅ F_{YZX}, F_{ZYX}	≅ ≅ F_{XZY}, F_{ZXY}	F_{XXY}	F_{YYX}	≅ ≅ F_{XZX}, F_{ZXX}	≅ ≅ F_{YZY}, F_{ZYY}	= ≅ F_{XYY}, F_{YXY}	= ≅ F_{YXX}, F_{XYX}	F_{YYY}	F_{XXX}

the designation of 'ordinary' and 'extraordinary' for biaxial crystals according to the discussion of sections 2.1.3. (ii) and 2.2.3. (ii).

We distinguish two groups of phase-matching cones for the biaxial crystals in accordance with the localization of the phase-matching directions with respect to the optical axis:

(i) The phase-matching directions which all have the same optical sign define a cone of type A (figures 2 and 5). Thus, the associated interactions have a polarization configuration of type 2o.e or 2e.o according to the criteria previously developed.

(ii) The phase-matching directions which change optical sign define a cone of type B (figure 5). The associated interactions change their polarization configuration on either side of the optical axis. We shall call 'interactions of type 2o.e 2e.o' the interactions which are of type 2o.e for the phase-matching direction located between the X axis and the optical axis in the (X, Z) plane. Thus, the configuration of polarization is of type 2e.o between the optical axis and the Z axis in the (X, Z) plane. The situation is reversed for the interactions of type 2e.o 2o.e.

NEGATIVE BIAXIAL CRYSTAL

	CONE TYPE A			CONE TYPE B		
λ (μm)	n_x	n_y	n_z	n_x	n_y	n_z
1	1,8200	1,8000	1,7700	1,8200	1,8000	1,7700
$\frac{1}{2}$	1,8700	1,8400	1,8200	1,8700	1,8400	1,8200
$\frac{1}{3}$	1,9200	1,8572	1,8267	1,9200	1,8470	1,8267

POSITIVE BIAXIAL CRYSTAL

	CONE TYPE A			CONE TYPE B		
λ (μm)	n_x	n_y	n_z	n_x	n_y	n_z
1	1,7400	1,7600	1,8200	1,7400	1,7600	1,8200
$\frac{1}{2}$	1,7700	1,8100	1,8700	1,7700	1,8100	1,8700
$\frac{1}{3}$	1,8000	1,8533	1,8989	1,7783	1,8533	1,9200

Phase-Matching relation : $\frac{n^+(\lambda_1)}{\lambda_1} + \frac{n^+(\lambda_2)}{\lambda_2} - \frac{n^-(\lambda_3)}{\lambda_3} = 0$

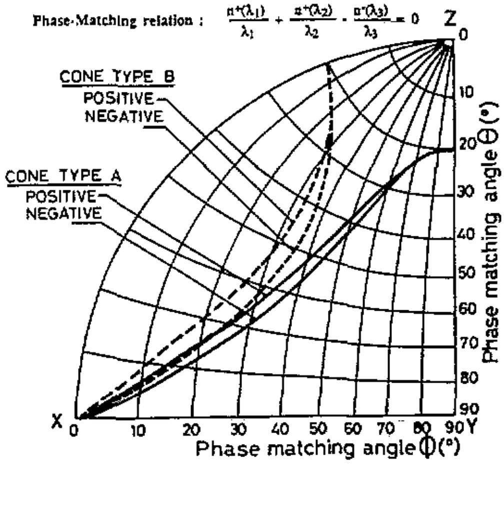


Figure 5. Refractive indices of biaxial crystals which leads to types A and B phase-matching 'cones' for the SFM of type I.

4.1. Interactions with a phase-matching cone of type A

This situation is directly comparable to that of the interactions in uniaxial crystals for which the phase-matching cones are always of type A.

The process is identical to the previous one. The birefringence and the dispersion in frequency of the refractive indices of the two typical biaxial crystals have been chosen so that the type I phase-matching directions of the SFM ($\lambda_1 = 1\,\mu$m, $\lambda_2 = 1/2\,\mu$m, $\lambda_3 = 1/3\,\mu$m) are identical for the two crystals. The data are given in figure 2. Furthermore these directions are near those of the two uniaxial crystals previously studied, in order to compare the field factors between uniaxial and biaxial classes. The field factors are calculated from the relations (12), (28), (29) and (30).

The 3-wave non-linear optical interactions of type 2o.e and of type 2e.o are described by twenty-seven field factors united in twenty-seven trigonometric functions. The field factors corresponding to the different interactions are given in tables 3, 7, 8 and 9.

For these two types of polarization configuration appear two groups of functions:

(i) The type 2o.e functions $(1, \overline{1})$, $(2, \overline{2})$, (3a, 3b), $(4\text{a}, 4\text{b}, \overline{4}\text{a}, \overline{4}\text{b})$, $(5, \overline{5})$ and the type 2e.o functions $(1, \overline{1})$, $(2\text{a}, 2\text{b}, \overline{2}\text{a}, \overline{2}\text{b})$, $(3, \overline{3})$, $(4\text{a}, 4\text{b}, \overline{4}\text{a}, \overline{4}\text{b})$, $(5\text{a}, 5\text{b}, \overline{5}\text{a}, \overline{5}\text{b})$, (6), $(\overline{6})$ concern the same coupling as those of the uniaxial classes and are called 'uniaxial functions'. Their angular variations are all the more similar to those of uniaxial classes

Table 7. Type (oee), (eeo) and (eoe) field factor tensors. · $F_{ijk}(\varphi) = 0$; ● $F_{ijk}(\varphi) \neq 0$, periodicity π; ⊙ $F_{ijk}(\varphi) \neq 0$, periodicity $\pi/2$. —— $F_{ijk}(\varphi) = F_{lmn}(\varphi)$; ------ $F_{ijk}(\varphi) = -F_{lmn}(\varphi)$; ═ $F_{ijk}(\varphi) \equiv F_{lmn}(\varphi)$; —•— $F_{ijk}(\varphi) = F_{lmn}(\varphi + \pi/2)$; ⇒ $F_{ijk}(\varphi) = -F_{lmn}(\varphi + \pi/2)$.

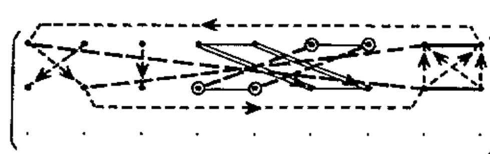
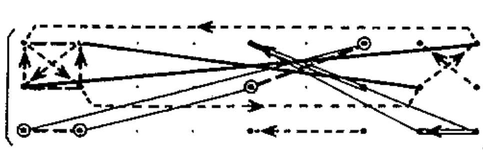
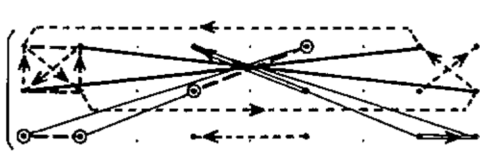
Interaction	Type 2 e.o F_{ijk} field factor tensor for uniaxial classes
	Type oee
SFM type I > 0, $\omega_3 = \omega_1 + \omega_2$; DFM type I < 0, $\omega_1 = \omega_3 - \omega_2$; $\omega_2 = \omega_3 - \omega_1$	XX YY ZZ YZ ZY XZ ZX XY YX; rows X, Y, Z: 
	Type eeo
SFM type II < 0, $\omega_3 = \omega_1 + \omega_2$; DFM type III < 0, $\omega_1 = \omega_3 - \omega_2$; DFM type II < 0, $\omega_2 = \omega_3 - \omega_1$	XX YY ZZ YZ ZY XZ ZX XY YX; rows X, Y, Z: 
	Type eoe
SFM type III < 0, $\omega_3 = \omega_1 + \omega_2$; DFM type II > 0, $\omega_1 = \omega_3 - \omega_2$; DFM type III > 0, $\omega_2 = \omega_3 - \omega_1$	XX YY ZZ YZ ZY XZ ZX XY YX; rows X, Y, Z: 

Table 8. Correspondence between the type 2o.e biaxial functions and the F_{ijk} coefficients associated with the different interactions of the family 2o.e.

Interactions	Optical sign	Type 2o.e or 2o.e2e.o Biaxial functions								
		6	$\bar{6}$	7 (7a, 7b)	$\bar{7}$ ($\bar{7}$a, $\bar{7}$b)	$\bar{8}$ ($\bar{8}$a, $\bar{8}$b)	8 (8a, 8b)	$\bar{9}$ ($\bar{9}$a, $\bar{9}$b)	9 (9a, 9b)	10
SFM $\omega_3 = \omega_1 + \omega_2$ Type I	<0	F_{YZZ}	F_{XZZ}	$\cong$ F_{XYZ}, F_{XZY}	$\cong$ F_{YXZ}, F_{YZX}	$\cong$ F_{XXZ}, F_{XZX}	$\cong$ F_{YYZ}, F_{YZY}	$\cong$ F_{ZYZ}, F_{ZZY}	$\cong$ F_{ZXZ}, F_{ZZX}	F_{ZZZ}
SFM $\omega_3 = \omega_1 + \omega_2$ Type II	>0	F_{ZZY}	F_{ZZX}	$\cong$ F_{ZYX}, F_{YZX}	$\cong$ F_{ZXY}, F_{XZY}	$\cong$ F_{ZXX}, F_{XZX}	$\cong$ F_{ZYY}, F_{YZY}	$\cong$ F_{ZYZ}, F_{YZZ}	$\cong$ F_{ZXZ}, F_{XZZ}	F_{ZZZ}
SFM $\omega_3 = \omega_1 + \omega_2$ Type III	>0	F_{ZYZ}	F_{ZXZ}	$\cong$ F_{ZXY}, F_{YXZ}	$\cong$ F_{ZYX}, F_{XYZ}	$\cong$ F_{ZXX}, F_{XXZ}	$\cong$ F_{ZYY}, F_{YYZ}	$\cong$ F_{ZZY}, F_{YZZ}	$\cong$ F_{ZZX}, F_{XZZ}	F_{ZZZ}
DFM $\omega_2 = \omega_3 - \omega_1$ Type III	<0	F_{ZYZ}	F_{ZXZ}	$\cong$ F_{ZXY}, F_{YXZ}	$\cong$ F_{ZYX}, F_{XYZ}	$\cong$ F_{ZXX}, F_{XXZ}	$\cong$ F_{ZYY}, F_{YYZ}	$\cong$ F_{ZZY}, F_{YZZ}	$\cong$ F_{ZZX}, F_{XZZ}	F_{ZZZ}
DFM $\omega_1 = \omega_3 - \omega_2$ Type I; DFM $\omega_2 = \omega_3 - \omega_1$ Type I	>0	F_{YZZ}	F_{XZZ}	$\cong$ F_{XZY}, F_{XYZ}	$\cong$ F_{YZX}, F_{YXZ}	$\cong$ F_{XZX}, F_{XXZ}	$\cong$ F_{YZY}, F_{YYZ}	$\cong$ F_{ZZY}, F_{ZYZ}	$\cong$ F_{ZZX}, F_{ZXZ}	F_{ZZZ}
DFM $\omega_1 = \omega_3 - \omega_2$ Type II	<0	F_{ZYZ}	F_{ZXZ}	$\cong$ F_{YXZ}, F_{ZXY}	$\cong$ F_{XYZ}, F_{ZYX}	$\cong$ F_{XXZ}, F_{ZXX}	$\cong$ F_{YYZ}, F_{ZYY}	$\cong$ F_{YZZ}, F_{ZZY}	$\cong$ F_{XZZ}, F_{ZZX}	F_{ZZZ}
DFM $\omega_1 = \omega_3 - \omega_2$ Type III; DFM $\omega_2 = \omega_3 - \omega_1$ Type II	>0	F_{ZZY}	F_{ZZX}	$\cong$ F_{YZX}, F_{ZYX}	$\cong$ F_{XZY}, F_{ZXY}	$\cong$ F_{XZX}, F_{ZXX}	$\cong$ F_{YZY}, F_{ZYY}	$\cong$ F_{YZZ}, F_{ZYZ}	$\cong$ F_{XZZ}, F_{ZXZ}	F_{ZZZ}

Table 9. Correspondence between the type 2e.o biaxial functions and the F_{ijk} coefficients associated with the different interactions of the family 2e.o.

		Type 2e.o or 2e.o 2o.e Biaxial functions								
	Optical			8		$\bar{9}$		9		
Interactions	sign	7	$\bar{7}$	8a	8b	$\bar{9}$a	$\bar{9}$b	9a	9b	10
SFM $\omega_3=\omega_1+\omega_2$ Type I	>0	F_{ZYY}	F_{ZXX}	≅ F_{ZXY}, F_{ZYX}		≅ F_{ZYZ}, F_{ZZY}		≅ F_{ZXZ}, F_{ZZX}		F_{ZZZ}
SFM $\omega_3=\omega_1+\omega_2$ Type II	<0	F_{YYZ}	F_{XXZ}	≅ F_{YXZ}, F_{XYZ}		≅ F_{ZYZ}, F_{YZZ}		≅ F_{ZXZ}, F_{XZZ}		F_{ZZZ}
SFM $\omega_3=\omega_1+\omega_2$ Type III	<0	F_{YZY}	F_{XZX}	≅ F_{YZX}, F_{XZY}		≅ F_{ZZY}, F_{YZZ}		≅ F_{ZZX}, F_{XZZ}		F_{ZZZ}
DFM $\omega_2=\omega_3-\omega_1$ Type III	>0	F_{YZY}	F_{XZX}	≅ F_{YZX}, F_{XZY}		≅ F_{ZZY}, F_{YZZ}		≅ F_{ZZX}, F_{XZZ}		F_{ZZZ}
DFM $\omega_1=\omega_3-\omega_2$ Type I; DFM $\omega_2=\omega_3-\omega_1$ Type I	<0	F_{ZYY}	F_{ZXX}	≅ F_{ZYX}, F_{ZXY}		≅ F_{ZZY}, F_{ZYZ}		≅ F_{ZZX}, F_{ZXZ}		F_{ZZZ}
DFM $\omega_1=\omega_3-\omega_2$ Type II	>0	F_{YZY}	F_{XZX}	≅ F_{XZY}, F_{YZX}		≅ F_{YZZ}, F_{ZZY}		≅ F_{XZZ}, F_{ZZX}		F_{ZZZ}
DFM $\omega_1=\omega_3-\omega_2$ Type III; DFM $\omega_2=\omega_3-\omega_1$ Type II	<0	F_{YYZ}	F_{XXZ}	≅ F_{XYZ}, F_{YXZ}		≅ F_{YZZ}, F_{ZYZ}		≅ F_{XZZ}, F_{ZXZ}		F_{ZZZ}

since n_x approaches n_y, that is the case with the chosen crystals. Thus, the functions are closed to those of figures 3 and 4.

(ii) The type 2o.e functions (6), ($\bar{6}$), (7a, 7b), ($\bar{7}$a, $\bar{7}$b), (8a, 8b), ($\bar{8}$a, $\bar{8}$b), (9a, 9b), ($\bar{9}$a, $\bar{9}$b) and (10) drawn in figure 6, and the type 2e.o functions (7, $\bar{7}$), (8a, 8b), (9a, 9b), ($\bar{9}$a, $\bar{9}$b) and (10) drawn in figure 7, are characteristic of the biaxial classes and are called 'biaxial functions'. The curves of the trigonometric functions with the suffices a and b are joined because of the low dispersion in frequency of the birefringence angles, as for the uniaxial crystals. These functions are all the smaller since the biaxial crystal 'tends' to a uniaxial crystal, that is n_x approaches n_y. This 'increase of the degree of symmetry', also concerns the $\chi^{(2)}$ tensor and involves additional equalities between certain elements of the tensor but not necessarily a decrease of their magnitude. Thus, for a given interaction, the elements of the tensor $\chi^{(2)}$ of a 'quasi uniaxial' biaxial crystal are weakly involved by the specific biaxial field factors. Nevertheless their global contribution can

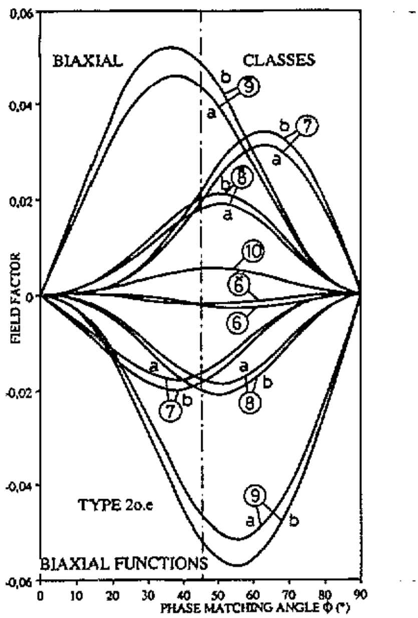


Figure 6. φ angular variation of type 2o.e biaxial field factors in biaxial crystals.

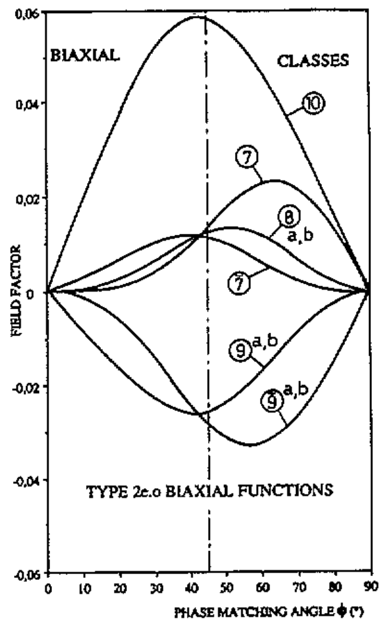


Figure 7. φ angular variation of type 2e.o biaxial field factors in biaxial crystals.

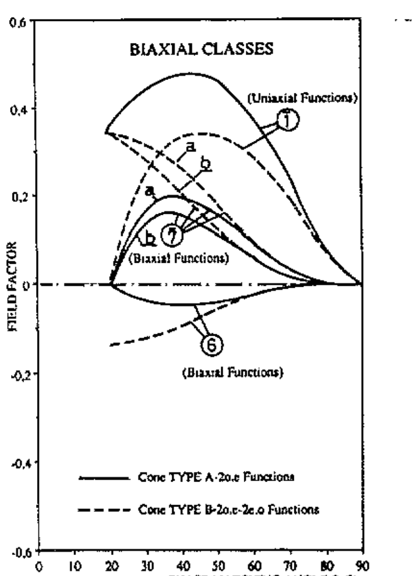


Figure 8. Types 2o.e and 2o.e 2e.o field factors which differ in the plane (X, Z).

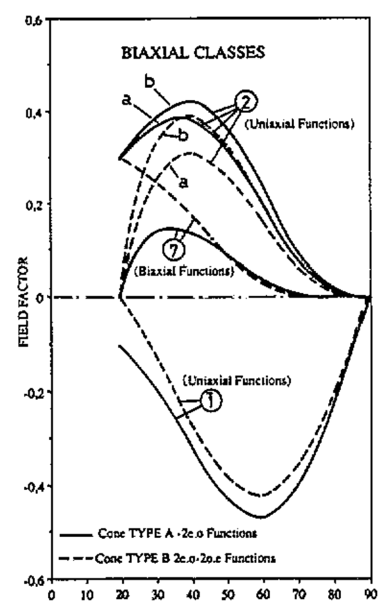


Figure 9. Types 2e.o and 2e.o 2o.e field factors which differ in the plane (X, Z).

Table 10. Intervening 2o.e and 2e.o uniaxial and biaxial functions for the five non-centrosymmetric biaxial classes of symmetry. The standard frame of orientation (x_1, x_2, x_3) is collinear to (X, Y, Z).

Biaxial acentric classes of symmetry	C_1	C_2 $(2\|x_3)$	C_2 $(2\|x_2)$
Intervening type 2o.e. or 2o.e.2e.o uniaxial and biaxial functions	$1, \bar{1}, 2, \bar{2}, 3, 4, \bar{4}, 5, \bar{5}, 6, \bar{6}, 7, \bar{7}, 8, \bar{8}, 9, \bar{9}, 10$	$1, \bar{1}, 3, 7, \bar{7}, 8, \bar{8}, 10$	$\bar{2}, 3, 4, 5, 6, 7, \bar{7}, 9, \bar{9}$
Intervening type 2e.o or 2e.o 2o.e uniaxial and biaxial functions	$1, \bar{1}, 2, \bar{2}, 3, \bar{3}, 4, \bar{4}, 5, \bar{5}, 6, \bar{6}, 7, \bar{7}, 8, 9, \bar{9}, 10$	$2, \bar{2}, 4, \bar{4}, 7, \bar{7}, 8, 10$	$1, 2, \bar{2}, 3, \bar{5}, 6, 8, \bar{9}$

Biaxial acentric classes of symmetry	C_s $(m \perp x_3)$	C_s $(m \perp x_2)$	C_{2v}	D_2
Intervening type 2o.e. or 2o.e 2e.o uniaxial and biaxial functions	$2, \bar{2}, 4, \bar{4}, 5, \bar{5}, 6, \bar{6}, 9, \bar{9}$	$1, \bar{1}, 2, \bar{4}, \bar{5}, \bar{6}, 8, \bar{8}, 9, 10$	$1, \bar{1}, 8, \bar{8}, 10$	$3, 7, \bar{7}$
Intervening type 2e.o or 2e.o 2o.e uniaxial and biaxial functions	$1, \bar{1}, 3, \bar{3}, 5, \bar{5}, 6, \bar{6}, 9, \bar{9}$	$1, \bar{3}, 5, \bar{6}, 7, \bar{7}, 9, 10$	$4, \bar{4}, 7, \bar{7}, 10$	$2, \bar{2}, 8$

be non-negligible in comparison with those of the uniaxial field factors, according to the sign of elements of the $\chi^{(2)}$ tensor.

As for the uniaxial classes, the two types of polarization configuration, 2o.e and 2e.o, are grouped together in three specific tensors. These tensors have twenty-seven non-zero elements. The relationships between elements which are valid for uniaxial crystals, are not strictly verified for the biaxial crystals, especially since n_x differs from n_y.

The intervening 2o.e and 2e.o functions are given in table 10 for the five non-centrosymmetric biaxial classes of symmetry. All the functions with the suffices 'a' and 'b' are not subject to the restrictions prevailing for the uniaxial classes. Thus, only the number of the functions of each couple appears in table 10: for example (8) for (8a) and (8b). The field factors corresponding to the different interactions are given in tables 3, 7, 8 and 9.

4.2. *Interactions with a phase-matching cone of type B*

The field factors of types 2o.e and 2e.o are calculated respectively for a typical negative crystal and for a typical positive crystal which respectively leads to a phase-matching cone of type A for the SFM of type I. They are compared with the field factors of the SFM of type I with a type B phase-matching cone in two typical crystals: one positive for the study of the type 2e.o 2o.e functions and another one negative for the study of the type

2o.e 2e.o functions. The refractive indices of these crystals and the phase-matching cones are given in figure 5.

In figures 8 and 9, only the trigonometric functions of types 2o.e and 2o.e 2e.o on the one hand and of types 2e.o and 2e.o 2o.e on the other hand which differ in the principal planes of the index surface are drawn.

5. Conclusion

The birefringence and dispersion in frequency of the refractive indices impose the polarizations of the coupled waves which compose the field factor tensor and which condition in part the non-linear interaction efficiency. The relations between the elements of the field factor tensor are governed by specific properties of symmetry, characteristic of the beam in interaction with the crystal. The design or use of a crystal for a given non-linear interaction requires taking into account the non-linear electric susceptibility and the field factor: the effective coefficient of an interaction can be nil even if the components of $\mathbf{F}$ and χ are non-zero. The classes of symmetry D_4 and D_6 for example forbid all interaction with a type 2o.e configuration of polarization and the classes C_{4v} and C_{6v} forbid the type 2e.o interactions and that for all phase-matching direction. However, the different attenuation factors which also depend on the linear optical properties, as well as the walk-off angle and the angular, temperature and spectral bandwidths, must be also considered for the calculation of the theoretical efficiency of interaction.

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